

# Electrical control of magnon propagation in multiferroic BiFeO<sub>3</sub> films

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The spin wave spectra of multiferroic BiFeO<sub>3</sub> films is calculated using a phenomenological Landau theory that includes magnetostatic effects. The lowest frequency magnon dispersion is shown to be quite sensitive to the angle between spin wave propagation vector and the Néel moment. Since electrical switching of the Néel moment has recently been demonstrated in this material, the sensitivity of the magnon dispersion permits direct electrical switching of spin wave propagation. This effect can be used to construct spin wave logical gates without current pulses, potentially allowing reduced power dissipation per logical operation.

One of the challenges of current research in microelectronic devices is the development of a fast logic switch with minimal power dissipation per cycle. Devices based on spin wave interference [1, 2] may provide an interesting alternative to conventional semiconductor gates by minimizing the need for current pulses. Recently, a spin wave NOT gate was demonstrated experimentally [1]. The device consisted of a current-controlled phase shifter made by a ferromagnetic (FM) film on top of a copper wire. The application of a current along the wire creates a local magnetic field on the film, leading to a phase shift of its spin waves.

In this letter we predict an effect that allows the design of similar spin wave devices without the need for external current pulses or applied time-dependent magnetic fields. We show that the dispersion of the lowest frequency spin-wave branch of a canted antiferromagnet depends strongly on the direction of spin wave propagation. This occurs because of the long-ranged (dipolar) interactions of the magnetic excitations, which creates a gap for spin waves propagating with non-zero projection along the Néel axis. This effect allows electrical control of spin waves in multiferroic materials that possess simultaneous ferroelectric (FE) and canted antiferromagnetic (AFM) order.

Our model is applicable to the prominent room temperature multiferroic BiFeO<sub>3</sub> (BFO) [3]. BFO films have homogeneous AFM order [4, 5], in contrast to the inhomogeneous (cycloidal) AFM order present in bulk BFO [6]. The canted AFM order in BFO films is constrained to be in the plane perpendicular to the FE polarization  $\mathbf{P}$ . Recently, Zhao *et al.* [7] demonstrated room temperature switching of the Néel moment  $\mathbf{L} = \mathbf{M}_1 - \mathbf{M}_2$  in BFO films after the orientation of the ferroelectric moment was changed electrically. As we show here, spin wave propagation along  $\mathbf{P}$  has high group velocity ( $\sim 10^5$  cm/s), in contrast to spin wave propagation along  $\mathbf{L}$  which has zero group velocity at  $\mathbf{k} = 0$ . Hence switching  $\mathbf{P}$  for a fixed spin wave propagation direction allows electrical control of the spin wave dispersion, which assuming some loss rate will effectively stop long-wavelength spin waves such as those created in [2].

Although a theory of AFM resonance for canted magnets was developed some time ago [8, 9], we are not aware of calculations of spin wave dispersion including magnetostatic effects. The electromagnon spectra for a ferromagnet with quadratic magnetoelectric coupling was discussed without magnetostatic effects in Ref. 10, and with magnetostatic effects in Ref. 11. Recently we developed a theory of spin wave dispersion in bulk BFO, a cycloidal (inhomogeneous) multiferroic [12]. The lowest frequency spin wave mode was shown to depend sensitively on the  $\mathbf{P}$  orientation because of the inhomogeneous nature of the antiferromagnetic order. Interestingly, we show here that BFO films with a homogeneous order display a similar effect, albeit due to a completely different physical reason: the magnetostatic effect.

Our calculation is based on a dynamical Ginzburg-Landau theory for the coupled magnetic and ferroelectric orders. We assume a model free energy given by

$$F = \frac{aP_z^2}{2} + \frac{uP_z^4}{4} + \frac{a_\perp(P_x^2 + P_y^2)}{2} - \mathbf{P} \cdot \mathbf{E} + \sum_{j=1,2} \left[ \frac{r\mathbf{M}_j^2}{2} + \frac{G\mathbf{M}_j^4}{4} + \frac{\alpha \sum_i (\nabla M_{ji})^2}{2} \right] + (J_0 + \eta P^2) \mathbf{M}_1 \cdot \mathbf{M}_2 + d\mathbf{P} \cdot \mathbf{M}_1 \times \mathbf{M}_2. \quad (1)$$

Here  $\mathbf{M}_j$  is the magnetization of one of the two sublattices  $j = 1, 2$ , and  $\mathbf{P}$  is a ferroelectric polarization. The coordinate system is such that  $\hat{\mathbf{z}}$  points along one of the cubic (111) directions in BFO. The exchange interaction  $J = (J_0 + \eta P^2)$  is assumed to have a quadratic dependence on  $P$  due to magnetostriction. The last contribution to Eq. (1) is a Dzyaloshinskii-Moriya (DM) interaction, with a DM vector given by  $d\mathbf{P}$ . Note that this changes sign under inversion symmetry, hence Eq. (1) is invariant under spatial inversion at a point in between the two sublattices.

The design of multiferroic materials with enhanced couplings of this type was recently discussed [13]. Although BiFeO<sub>3</sub> has no inversion center, its crystal structure is quite close to an inversion-symmetric one, and the above free energy is derived by assuming that both the

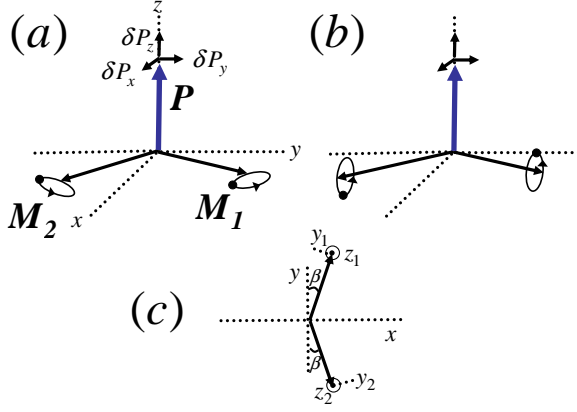


FIG. 1: Spin and polarization waves in a canted multiferroic, such as a BiFeO<sub>3</sub> film. The sublattice magnetizations  $\mathbf{M}_1$ ,  $\mathbf{M}_2$  lie in the plane perpendicular to the FE polarization  $\mathbf{P}$ . Fluctuations  $\delta\mathbf{P}$  denote polar phonons associated to vibrations of the FE moment. (a) Depicts the low frequency (soft) spin wave mode. (b) High-frequency (gapped) mode. The dots in the circle denote the position of the spins one quarter cycle later. The soft mode leaves the canting angle  $\beta$  invariant, while the gapped mode modulates  $\beta$ . (c) Coordinate system.

DM vector and polarization  $\mathbf{P}$  are associated with the same distortion of the lattice. An alternative model for BiFeO<sub>3</sub> assumes the DM vector to be independent of  $\mathbf{P}$  [14], i.e., requires Eq. (1) be invariant under spatial inversion at a point on top of one of the magnetic ions. Later we will discuss the implications of this alternative assumption for the electromagnon spectra, and show how optical experiments may determine which model is appropriate.

The free energy is minimized by a homogeneous ferroelectric and antiferromagnetic state, with FE moment (at  $\mathbf{E} = 0$ ) given by  $\mathbf{P} = P_0\hat{z}$ , with  $P_0^2 = \frac{-a}{u} + \mathcal{O}(d^3)$ . The magnetic moments are perpendicular to  $\mathbf{P}$ ,

$$\mathbf{M}_{01} = M_0 (\sin \beta \hat{x} + \cos \beta \hat{y}), \quad (2a)$$

$$\mathbf{M}_{02} = -M_0 (-\sin \beta \hat{x} + \cos \beta \hat{y}), \quad (2b)$$

with canting angle  $\beta$  and magnetization  $M_0$  determined by  $\tan \beta = (dP_0)/(\tilde{J} + J)$ , and  $M_0^2 = (\tilde{J} - r)/G$ , with  $\tilde{J}^2 = (dP_0)^2 + J^2$ . Below the Curie and Néel temperatures we have  $a < 0$ , and  $J > -r > 0$  respectively.

Small oscillations away from the ground state are described by the Landau-Lifshitz equations,

$$\frac{\partial \mathbf{M}_i}{\partial t} = \gamma \mathbf{M}_i \times \frac{\delta F}{\delta \mathbf{M}_i}, \quad (3)$$

where  $\gamma$  is a gyromagnetic ratio. A corresponding set of equations is written for  $\mathbf{P}$  in order to describe the high frequency optical phonon spectra. Keeping only the lowest order in the deviations  $\delta \mathbf{M}_i$  and  $\delta \mathbf{P}$ , and focusing on the low frequency magnetic oscillations we seek plane wave

solutions of the type

$$\mathbf{M}_i = \mathbf{M}_{0i} + \delta \mathbf{M}_i e^{i(\mathbf{k} \cdot \mathbf{r} - \omega t)}, \quad \mathbf{P} = P_0 \hat{z} + \delta \mathbf{P} e^{i(\mathbf{k} \cdot \mathbf{r} - \omega t)}. \quad (4)$$

From Eq. (3) we see that  $\delta \mathbf{M}_i$  must be perpendicular to  $\mathbf{M}_i$ . Hence we may reduce the number of variables by using a parametrization for  $\delta \mathbf{M}_i$  shown in Fig. 1(c), with further definitions  $Y = y_1 + y_2$ ,  $Z = z_1 + z_2$ ,  $y = y_1 - y_2$ ,  $z = z_1 - z_2$ .

From Maxwell's equations we see that any macroscopic wave producing nonzero fluctuations of  $\delta \mathbf{M} = \delta \mathbf{M}_1 + \delta \mathbf{M}_2$  must induce an AC magnetic  $\mathbf{h}$  field. In the magnetostatic approximation this is obtained from  $\nabla \cdot \mathbf{h} = -4\pi \nabla \cdot \delta \mathbf{M}$  and  $\nabla \times \mathbf{h} \approx 0$ . The latter assumes the time variations are negligible in Maxwell's equations, which is a good approximation for spin waves provided  $k \gg \omega_{\text{AFM}}/c$ , with  $c$  the speed of light. For a canted AFM this is a good approximation provided the domain sizes are smaller than a few centimeters. The self induced field is therefore

$$\mathbf{h} = -4\pi (\delta \mathbf{M} \cdot \hat{\mathbf{n}}) \hat{\mathbf{n}}, \quad (5)$$

where  $\hat{\mathbf{n}}$  is a propagation direction for the spin waves,  $\mathbf{k} = k\hat{\mathbf{n}}$ . The self-induced field contributes a term  $2\pi(\delta \mathbf{M} \cdot \hat{\mathbf{n}})^2$  to the free energy, tending to increase the spin wave frequencies whenever the quantity  $\delta \mathbf{M} = (-\cos(\beta)y, \sin(\beta)Y, Z)$  has a finite projection along  $\hat{\mathbf{n}}$ .

In the magnetostatic approximation the linearized equations of motion are obtained by substituting Eqs. (4)-(5) into Eq. (3), and using the explicit expressions for  $\tan \beta$  and  $M_0$ . After some algebra the Landau-Lifshitz equations become

$$-i\tilde{\omega}Y + (\tilde{J} + J + \alpha k^2)Z - 2h_z = -2d' \cos \beta \delta P_x, \quad (6a)$$

$$\alpha k^2 Y + i\tilde{\omega}Z - 2\sin \beta h_y = -4\eta' \sin 2\beta \delta P_z, \quad (6b)$$

$$i\tilde{\omega}z + (2\tilde{J} + \alpha k^2)y + 2\cos \beta h_x = -2d' \cos 2\beta \delta P_z, \quad (6c)$$

$$(\tilde{J} - J + \alpha k^2)z - i\tilde{\omega}y = -2d' \sin \beta \delta P_y, \quad (6d)$$

where we defined  $\tilde{\omega} = \omega/(\gamma M_0)$ ,  $d' = dM_0$ , and  $\eta' = \eta P_0 M_0$ .

Consider the pure spin waves in the limit  $\delta \mathbf{P} \rightarrow 0$ . This case may be solved analytically, because the system of four equations decouples into two independent sets of equations on the variables  $(Y, Z)$  and  $(y, z)$ . The former is a low frequency mode, because it corresponds to spin vibrations that leaves the canting angle  $\beta$  unchanged [the spins vibrate in phase, see Fig 1(a)]. The latter corresponds to spin vibrations half-cycle out of phase, leading to modulations of  $\beta$ , and a high frequency gap equal to the DM interaction  $dP_0$  [Fig. 1(b)]. Neglecting terms to second order in  $(dP_0)/J$ , we may get an analytical expression for the low frequency mode,

$$\tilde{\omega}^2(\mathbf{k}) \approx 2J \left( 1 + \frac{4\pi}{J} n_z^2 \right) \alpha k^2 + \frac{4\pi(dP_0)^2}{J} n_y^2. \quad (7)$$

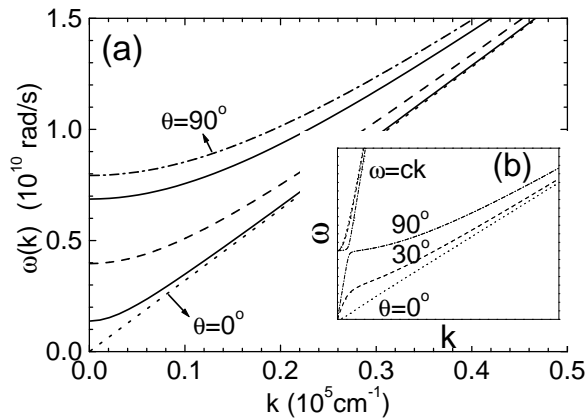


FIG. 2: (a) Low frequency magnetostatic spin wave dispersion for a BiFeO<sub>3</sub> film, for propagation angles  $\theta = 0^\circ$  (propagation along the electric polarization direction  $\hat{z}$ ),  $10^\circ$ ,  $30^\circ$ ,  $60^\circ$ ,  $90^\circ$  (propagation along the Néel direction  $\hat{y}$ ). The high frequency mode (not shown) has a gap equal to the Dyzyaloshinskii-Moriya coupling ( $5 \times 10^{10}$  rad/s), and is nearly isotropic with respect to the direction of spin wave propagation. (b) Dispersion including electrodynamical effects in the  $k < \omega/c$  region. Note the relationship between the magnetostatic gap in (a) and the photon-magnon anticrossing in (b).

This dispersion is anisotropic with respect to the polarization ( $\hat{z}$ ) axis: For  $\mathbf{k}$  along the  $x-z$  plane, we have a truly gapless mode to all orders in  $dP_0/J$ , with  $\tilde{\omega} \approx \sqrt{2J\alpha}k$ . For  $\mathbf{k}$  along  $\hat{y}$  we find a gap equal to a fraction of the DM interaction,  $\approx \sqrt{4\pi/J}(dP_0)$ . This gap is a result of the *magnetostatic correction in the presence of DM weak ferromagnetism*.

The physical origin of the magnetostatic gap is found by noting that  $\delta\mathbf{M}$  for a pure soft mode  $Y, Z \neq 0, y, z = 0$  as  $k \rightarrow 0$  is approximately given by a rigid rotation around the  $\hat{z}$  axis. In this limit,  $\delta\mathbf{M}$  points exclusively along  $\hat{y}$ , hence only propagation with some projection in this direction leads to a gap.

A small anisotropy is also found for the high frequency mode ( $y, z \neq 0, Y = Z = 0$ ). For example, when  $\hat{\mathbf{k}} \parallel \hat{\mathbf{x}}$  the high frequency mode gap increases to  $dP_0\sqrt{1 + 4\pi/J}$ .

We calculated the coupled spin and polarization wave spectra solving the full set of Eqs. (6a)-(6d) numerically, with parameters extracted from experiment [3, 4, 5]. The low frequency spin wave branch within the magnetostatic approximation is shown in Fig. 2(a). The inset [Fig. 2(b)] shows the low frequency spectra beyond the magnetostatic approximation, including electrodynamical corrections (For numerical convenience the speed of light was rescaled to  $10^6$  cm/s). Note the anticrossing of the spin wave modes with the photon dispersion  $\omega = ck$ , and the orientation dependence of the photon gap. As expected, we see that the strict  $k \rightarrow 0$  limit has no orientation dependence. We emphasize that the latter low  $k$  limit is only observable for domain sizes of one cm or larger. The magnetostatic propagation anisotropy discussed in

this work arises precisely because the spin waves travel with finite  $k > \omega/c$ .

Finally, we discuss the selection rules for the excitation or detection of magnon modes using an AC electric field. From inspecting the right hand side of Eqs. (6a) and (6b) we see that the low frequency magnon may be excited electrically by the application of an AC field in the  $x$  or  $z$  direction. The former has a strong response in the presence of linear magnetoelectric effect ( $d \neq 0$ ), while the latter has a weak response ( $\propto \sin\beta$ ) due to magnetostriction.

The high frequency magnon ( $x, y$ ) has a dielectric response only in the presence of the linear magnetoelectric effect, as seen in Eqs. (6c) and (6d). This mode responds to electric fields in the  $y-z$  plane, with the  $z$  direction response larger by a factor of  $\cos 2\beta/\sin\beta \sim 2J/dP_0 \gg 1$ . The presence or absence thereof of this electromagnon using an optical probe may be used to discern whether the DM vector is linear in  $P$  as proposed e.g. in [15] or if it is independent of  $P$  as suggested in [14].

In conclusion, we predicted a magnetostatic gap anisotropy for the propagation of spin waves in a canted antiferromagnet. This effect may allow the electrical switching of magnons in multiferroic materials such as BiFeO<sub>3</sub> films.

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